V|vwhp dwlf vwxg| ri wkhwhuqdu| ilvvlrq rigliihuhqwFp dqg Fi lvrwrshv

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Brix meeting
SCK•CEN Mol, Belgium
6-9th April 2008

UNIVERSITEIT GENT

irmm Institute for Reference Materials and Measurements

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- Introduction
- Experimental conditions
  - location
  - samples
  - detection system
- Analysis & Results
  - particle identification
  - Cm isotopes
  - Cf isotopes
- Discussion: systematic study
- Conclusions and outlook

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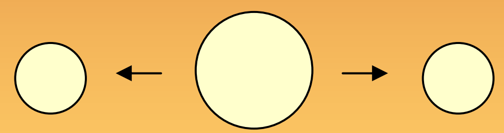
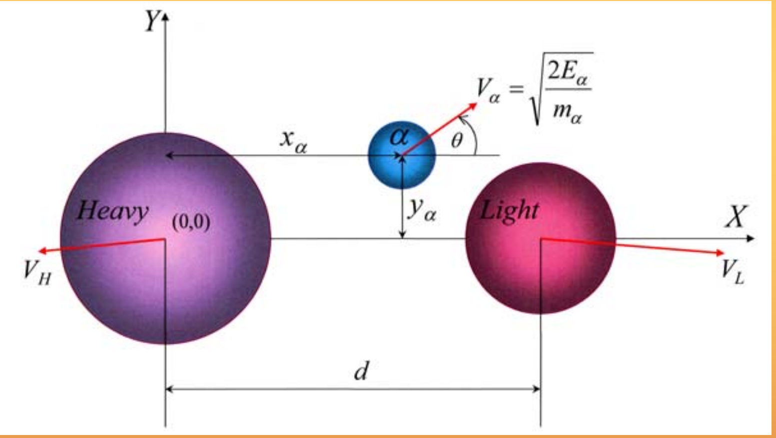
- Fission: a binary process

  Two heavy fragments are formed when the fissioning nucleus splits



- Ternary fission:
  three particles are formed

  discovered in 1946



Light Charged Particle (LCP) accompanied fission

$1 \leq Z \leq 10$ 2 to 4 times every thousand fission events

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Why do we measure ternary particles?

- Applied research

  Important source of helium and tritium gas production
  in nuclear reactors and used fuel elements

  Accurate data requested by nuclear industry and safety specialists

- Fundamental research

  Improving our understanding of the ternary particle emission

  Ternary particles are emitted in space and time close to the scission point
  → information on the fission process itself

Aim of this work:

- Enlarge the available databases → yields and energy distributions
- Search for systematics in these parameters
- Examine influence of the excitation energy

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Emission probabilities or yields

- most common: ternary alpha particles
  - → Long Range Alpha (LRA) particles (↔ radioactive alpha decay)
- tritons (t) and ^{6}He particles

Energy distributions

- Gaussian in shape (central limit theorem)
- Average energy and Full Width at Half Maximum (FWHM)
- Difficult to measure low energy part
  - → Threshold due to thickness of detector and protecting foils

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Influence excitation energy on emission probabilities

Comparing for the same compound nucleus:

- Spontaneous fission (SF)

  E_{exc} = 0 MeV

- Neutron induced fission (n_{th},f)

  E_{exc} = neutron binding energy ≈ 6-7 MeV

- ^{243}Cm(n_{th},f) & ^{244}Cm(SF)
- ^{245}Cm(n_{th},f) & ^{246}Cm(SF)
- ^{247}Cm(n_{th},f) & ^{248}Cm(SF)
- ^{249}Cf(n_{th},f) & ^{250}Cf(SF)
- ^{251}Cf(n_{th},f) & ^{252}Cf(SF)

Cm and Cf isotopes

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Location of the measurements

- Spontaneous fission
  - Institute for Reference Materials and Measurements (IRMM)
  - Geel (Belgium)
- Neutron induced fission
  - Institut Laue-Langevin (ILL)
  - Grenoble (France)

- PF1b neutron guide
- 76 m length
- High flux reactor: 10^9-10^{10} n/cm^2/s

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Samples

The amount of this exotic material is limited, and it is difficult to get these samples

^{243}Cm & ^{244}Cm

- Prepared at the Russian Federal Nuclear Center (RFNC)
- Spot curium oxide (Ø = 15 mm) deposited on Al-foil
- Activity ^{243}Cm = 3.7 MBq
  ^{244}Cm = 53.7 MBq

$T_{1/2,\alpha}(^{243}\mathrm{Cm}) = 29.10$ y → Production of ^{239}Pu formed by alpha decay
Correction had to be made!

$T_{1/2,SF}(^{244}\mathrm{Cm}) = (1.32 \pm 0.02) \times 10^{7}$ y; $T_{1/2,\alpha} = 18.10$ y

Unfavourable ratio to measure spontaneous fission, and dealing with very active source

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Samples

- ^{251}Cf & ^{252}Cf
  - Prepared at the Institut fur Kernchemie in Mainz
  - Spot californium oxide (Ø = 4 mm) deposited on Ti-foil
  - Activity ^{251}Cf = 10.8 MBq
    ^{252}Cf = 70 kBq
- ^{249}Cf & ^{250}Cf
  - Prepared at the Lawrence Berkeley National Laboratory (LBNL)
  - Spot californium oxide (Ø = 6 mm) deposited on Ti-foil
  - Activity ^{249}Cf = 0.9 MBq
    ^{250}Cf = 4 MBq

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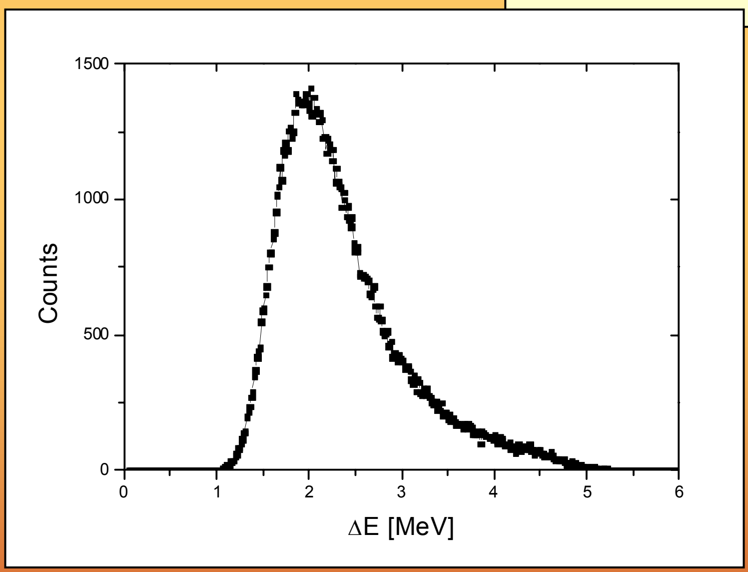
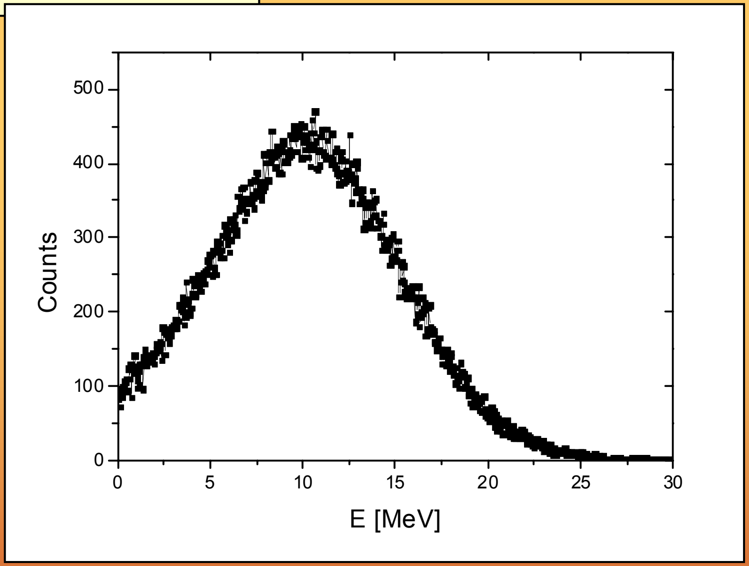
Detection system

- Various methods can be used for detection of the particles
  simple / not selective e.g. single surface barrier detector
- Need for particle identification

ΔE-E telescope detectors





ΔE-signal ~ energy deposited by the ternary particle traversing the first detector

E-signal ~ the remaining ternary particle energy

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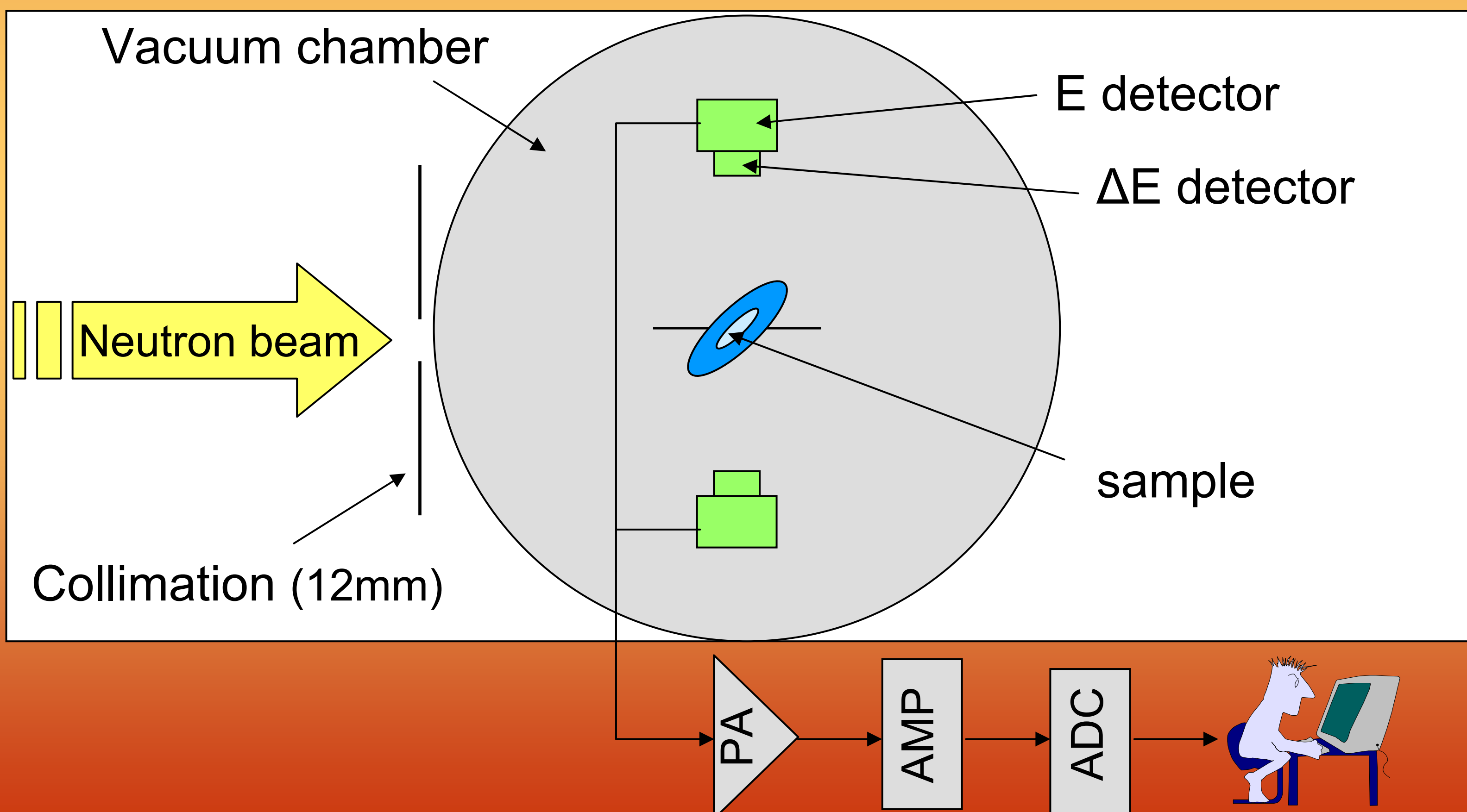
Detection system: neutron induced fission

Combination of two silicon surface barrier detectors



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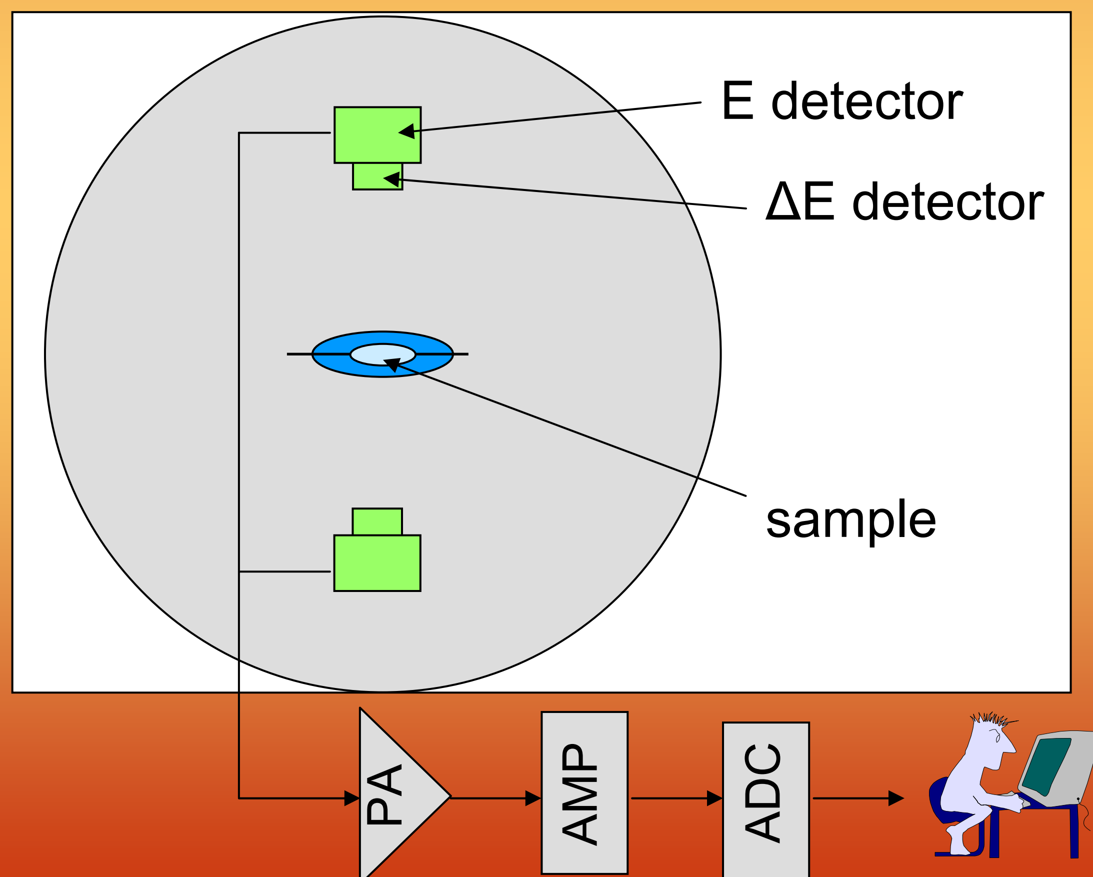
Detection system: spontaneous fission

Two different configurations

- Vacuumchamber with a combination of two silicon surface barrier detectors



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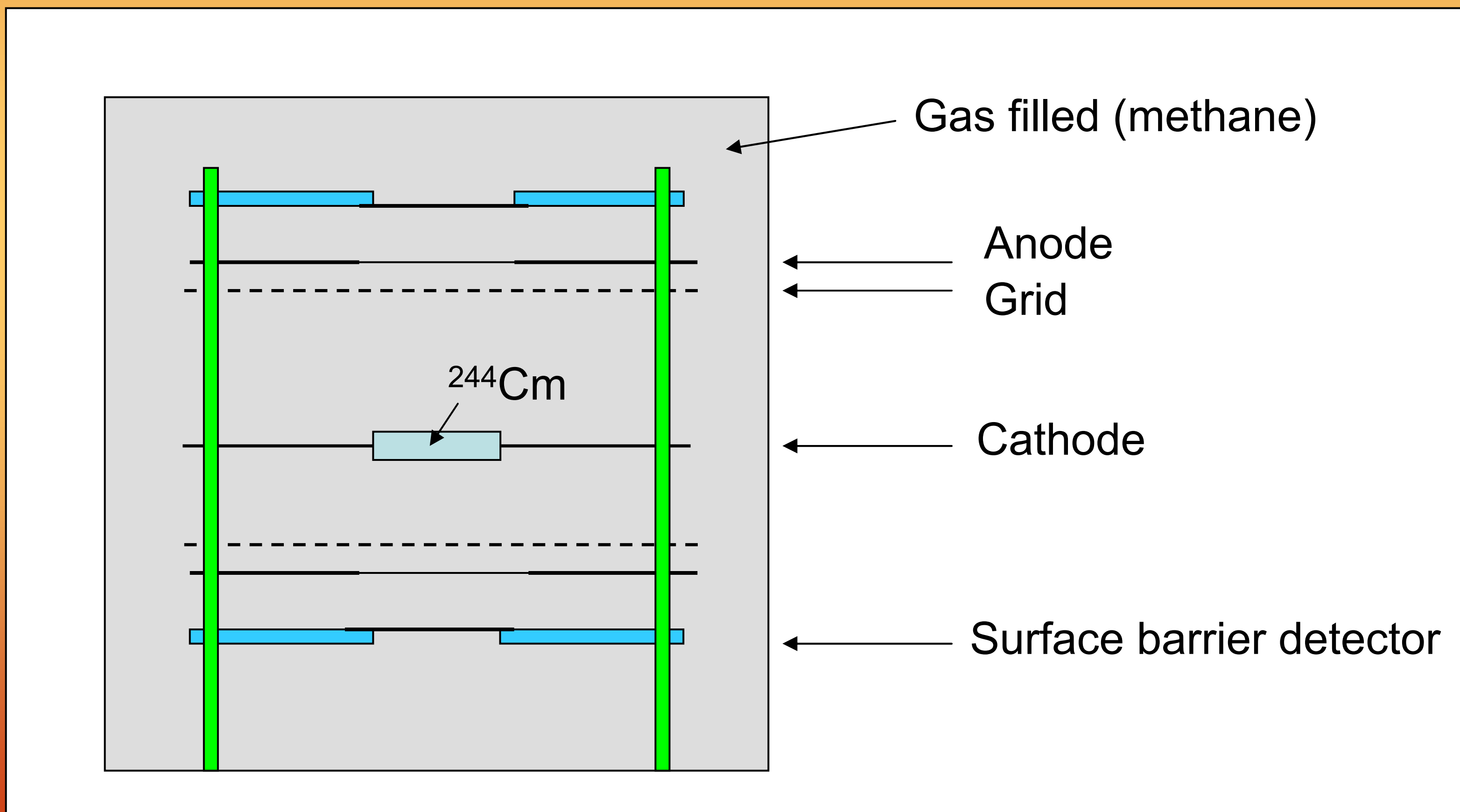
Detection system: spontaneous fission

- Ionisation chamber (ΔE) in combination with a surface barrier detector (E)



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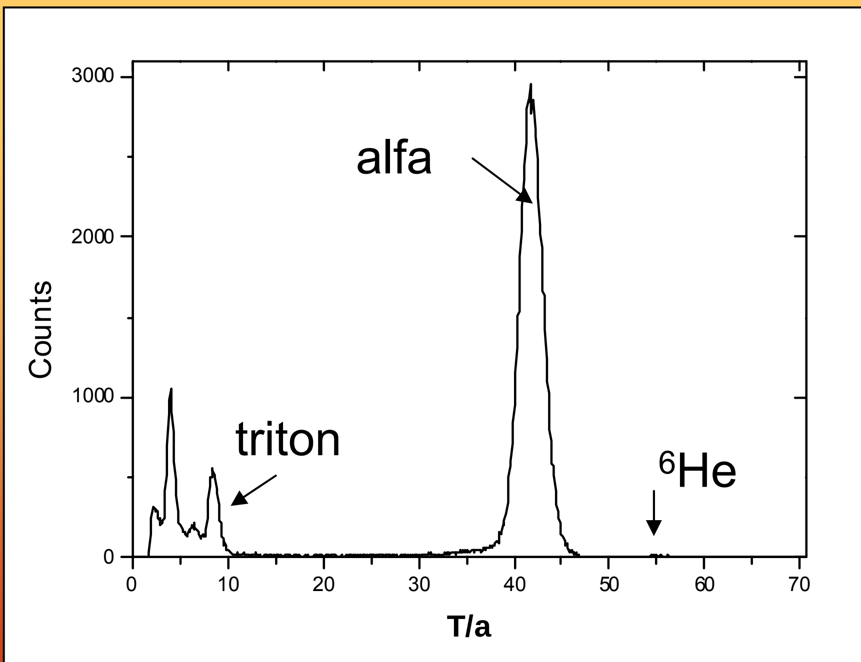
Particle identification

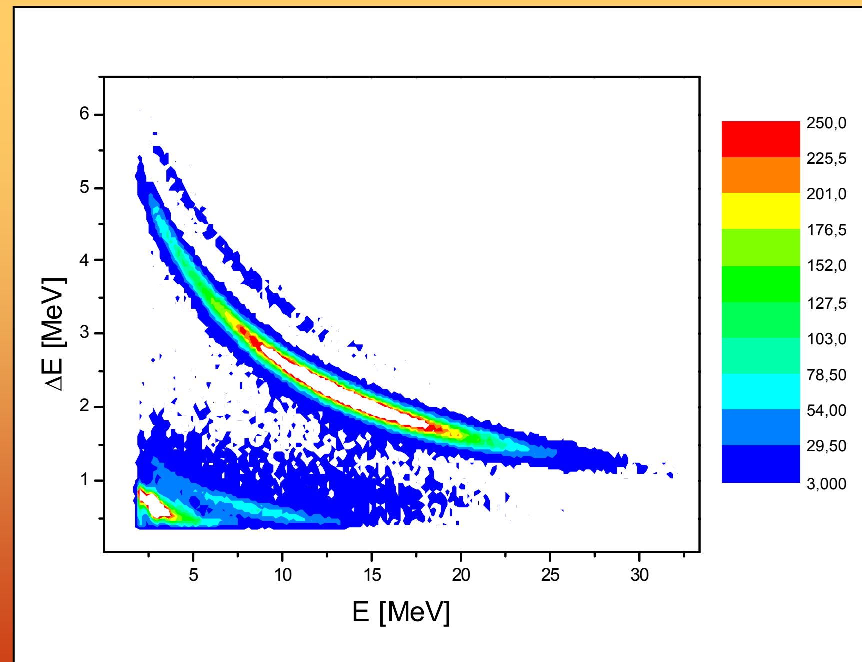
(Goulding)

Based on the difference in energy loss of different particles in the same material

$$T/a = (E + \Delta E)^{1.73} - E^{1.73}$$

(T = thickness of ΔE detector, a particle and material specific constant)





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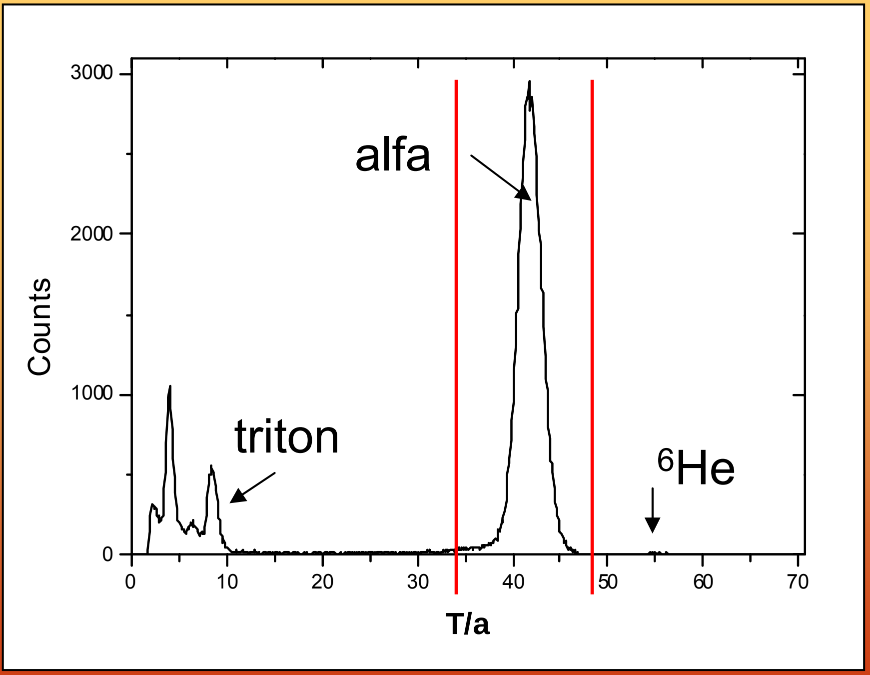

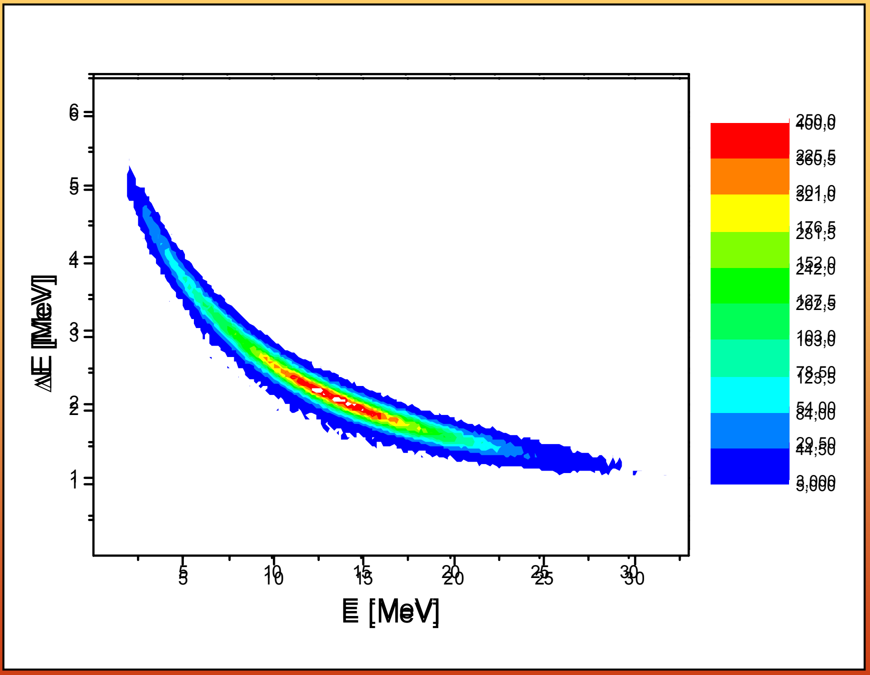

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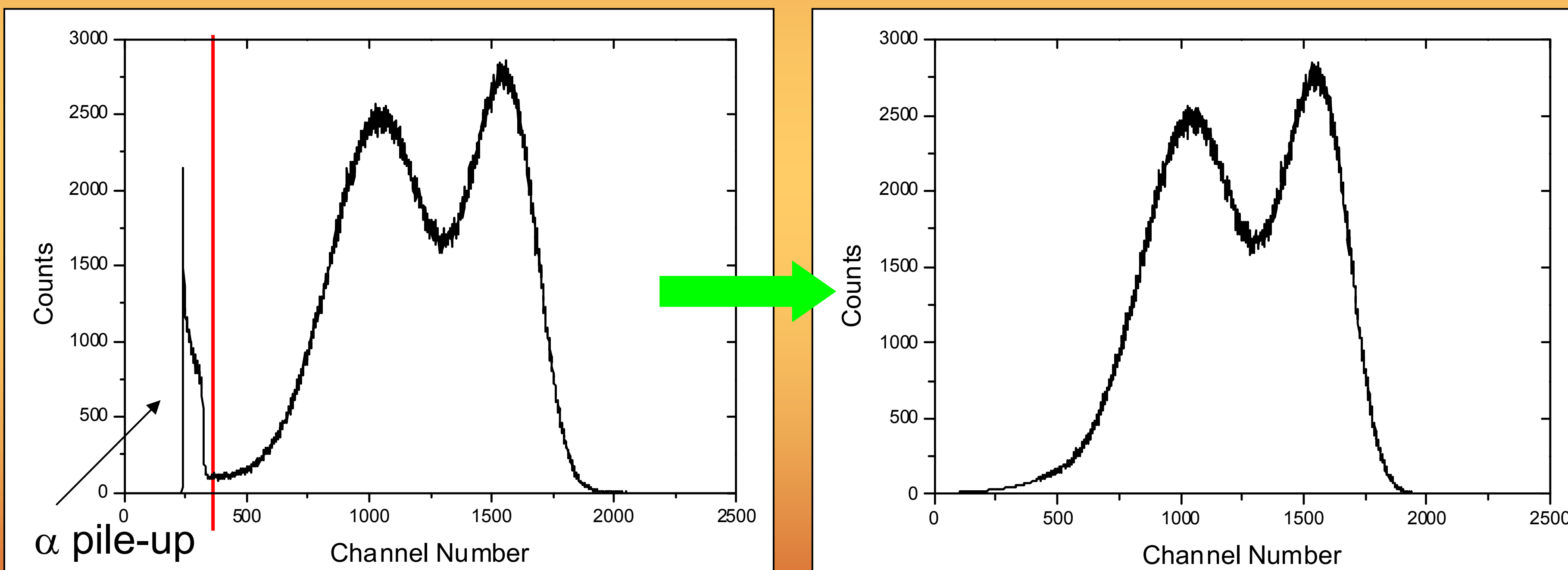
Binary Fission (B)

First step: measurement only with E detector, ΔE replaced by a dummy, detection of the fission fragments



227.16 ± 1.32 B/s

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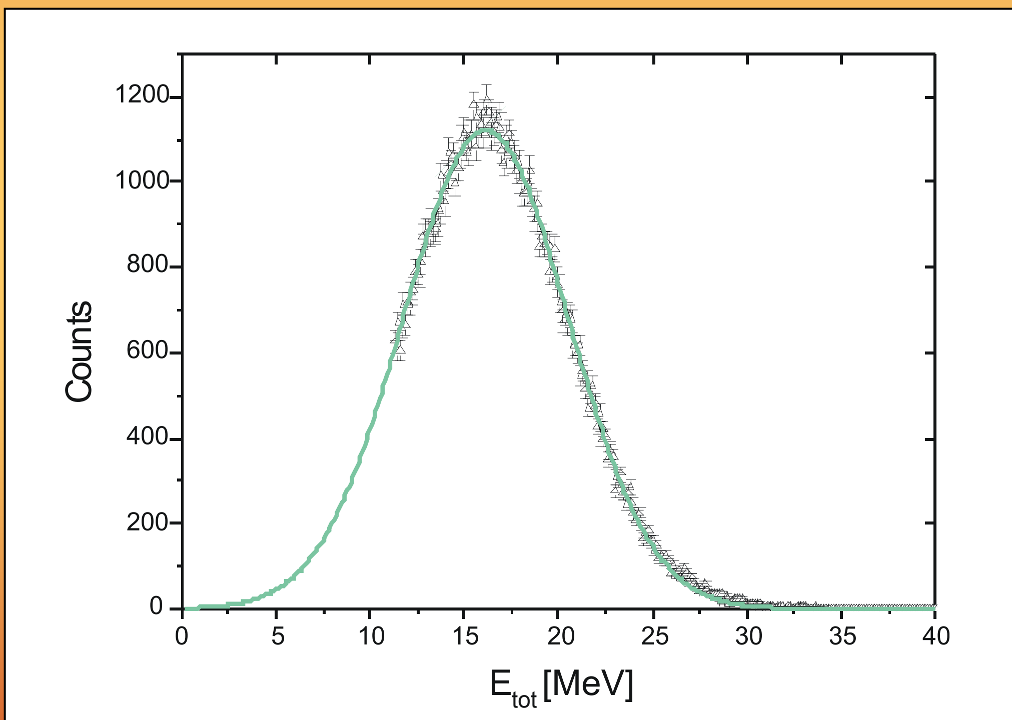
Second step: measurement with ΔE-E detector,
detection of the ternary particles

LRA-particles: Gaussian fit from 12.5 MeV



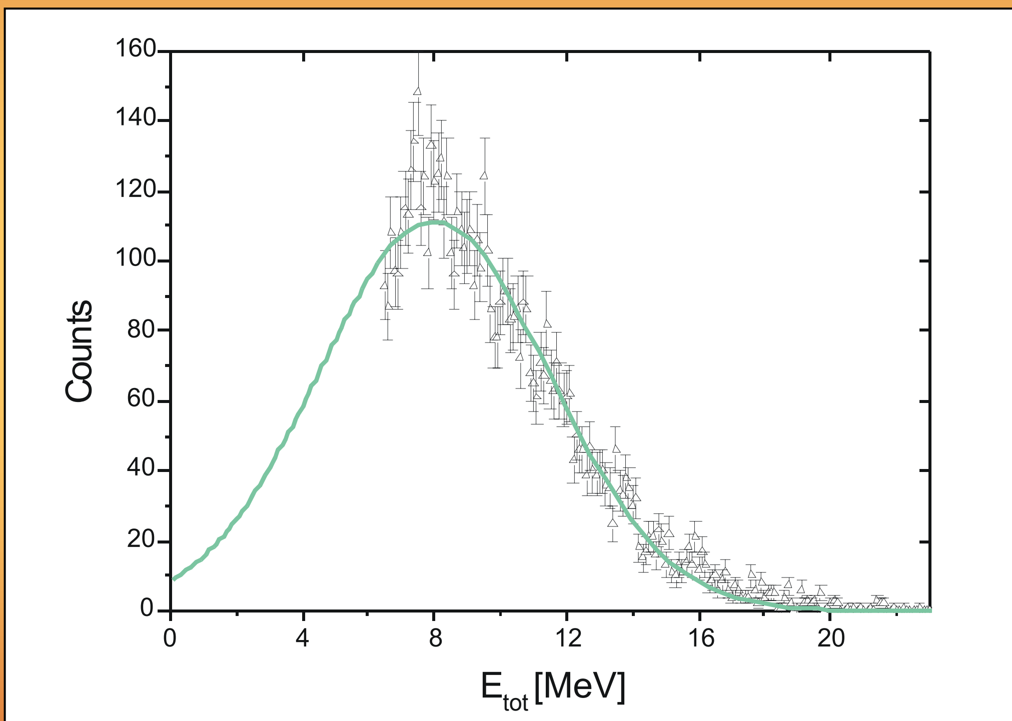
$E = 16.14 \pm 0.06$ MeV

$FWHM = 10.32 \pm 0.11$ MeV

Combining both results: $LRA/B = (2.43 \pm 0.08)\times10^{-3}$

Dqdo|vlv) Uhvxow ^{576}Fp

Tritons t: Gaussian fit to all data points



E = 8.15 ± 0.31 MeV

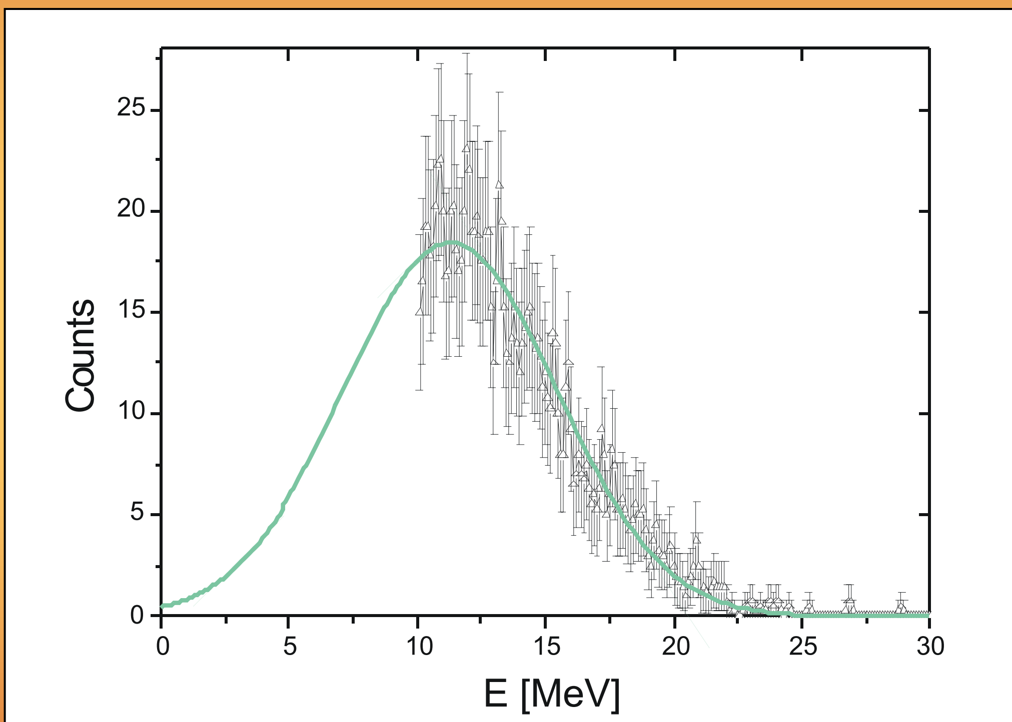
FWHM = 8.13 ± 0.47 MeV

Combining the results: t/LRA = (8.43 ± 1.10)%

t/B = $(1.77 \pm 0.39) \times 10^{-4}$

Dqdo|vlv) Uhvxow ^{576}Fp

^{6}He-particles: Gaussian fit from 10 MeV



E = 11.28 ± 0.53 MeV

FWHM = 9.79 ± 0.71 MeV

Combining the results: ^{6}He/LRA = (1.65 ± 0.32)%

^{6}He/B = $(4.04 \pm 0.81) \times 10^{-5}$

Dqdo|vlv) Uhvxow ^{577}Fp

Part I: Determination of LRA/B with ionisation chamber

$$LRA/B = (3.16 \pm 0.09)\times 10^{-3}$$

Part II: Determination of ternary particles with vacuum chamber

LRA-particles:

E = 15.99 ± 0.08 MeV

FWHM = 9.99 ± 0.29 MeV

Tritons t:

E = 8.05 ± 0.29 MeV

FWHM = 7.89 ± 0.48 MeV

➡ t/LRA = (6.75 ± 1.43)%; t/B = $(1.91 \pm 0.41)\times 10^{-4}$

^{6}He-particles:

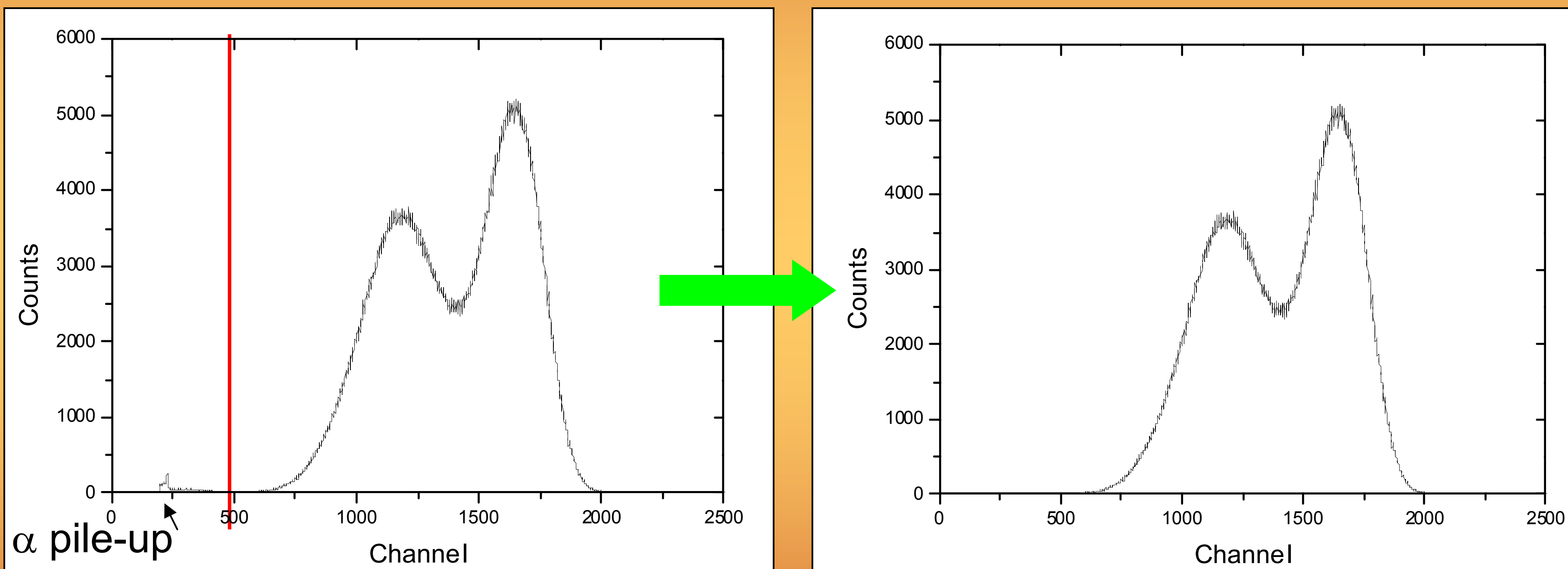
E = 11.22 ± 0.74 MeV

FWHM = 9.90 ± 1.14 MeV

➡ ^{6}He/LRA = (2.05 ± 0.52)%; ^{6}He/B = $(6.44 \pm 1.64)\times 10^{-5}$

Dqdo|vlv) Uhvxow 57<Fi

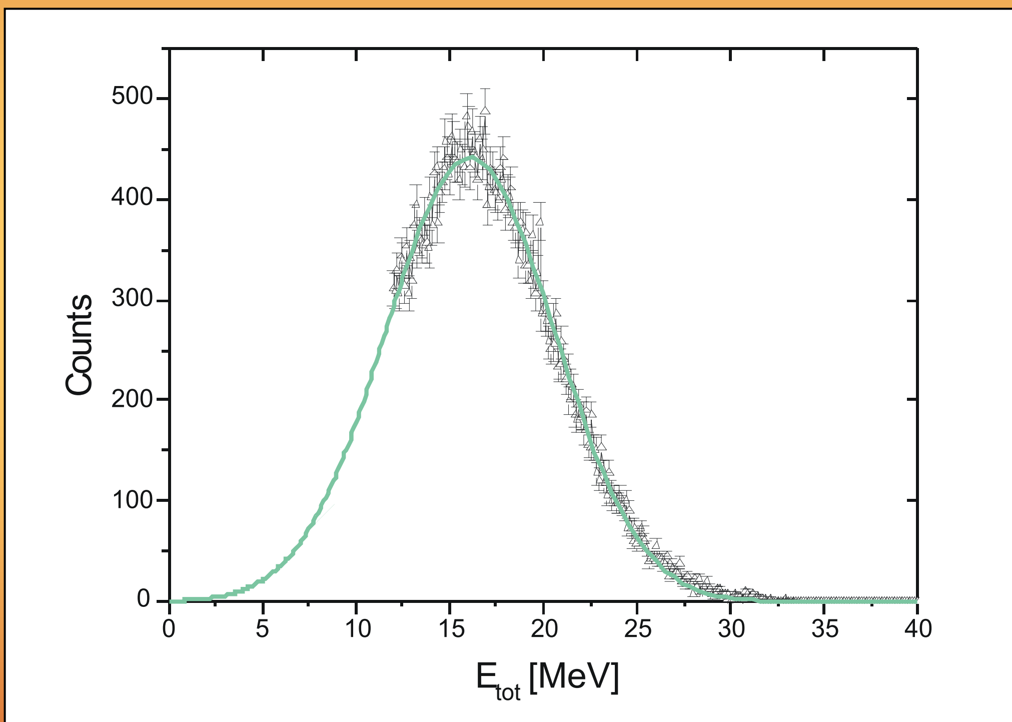
Binary Fission (B)



326.68 ± 1.79 B/s

Dqdo|vlv) Uhvxow 57<Fi

LRA-particles: Gaussian fit from 12.5 MeV



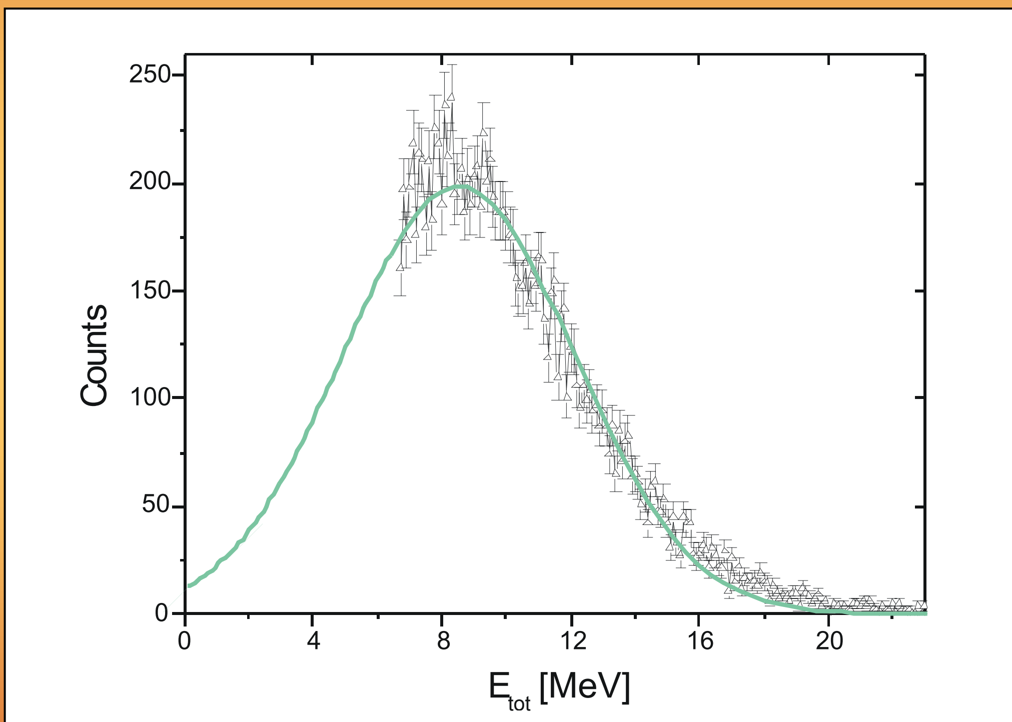
E = 16.09 ± 0.18 MeV

FWHM = 10.64 ± 0.27 MeV

Combining both results: LRA/B = (2.77 ± 0.11)x10^{-3}

Dqdo|vlv) Uhvxow   57<Fi

Tritons t: Gaussian fit to all data points



E = 8.52 ± 0.26 MeV
FWHM = 8.47 ± 0.56 MeV

Combining the results:

t/LRA = (7.9 ± 1.2)%
t/B = (2.20 ± 0.35)x10^{-4}

Dqdo|vlv) Uhvxow 584Fi2583Fi

Results obtained from the measurement of ^{251}Cf

LRA-particles:

E = 15.89 ± 0.12 MeV

FWHM = 10.57 ± 0.18 MeV

LRA/B = (2.40 ± 0.14)x10^{-3}

Tritons t:

E = 8.50 ± 0.10 MeV

FWHM = 8.37 ± 0.16 MeV

→ t/LRA = (9.00 ± 0.93)%; t/B = (2.17 ± 0.29)x10^{-4}

Measurement on ^{250}Cf

Experiment is just started
too early for results
at first sight everything looks fine

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t and α emission studied under the same experimental conditions
(C. Wagemans et al.)

Neutron induced fission

^{229}Th, ^{233,235}U, ^{237}Np, ^{239,241}Pu,
^{241,243}Am, ^{243,245,247}Cm, ^{249,251}Cf

Spontaneous fission

^{238,240,242,244}Pu, ^{244,246,248}Cm,
^{250,252}Cf

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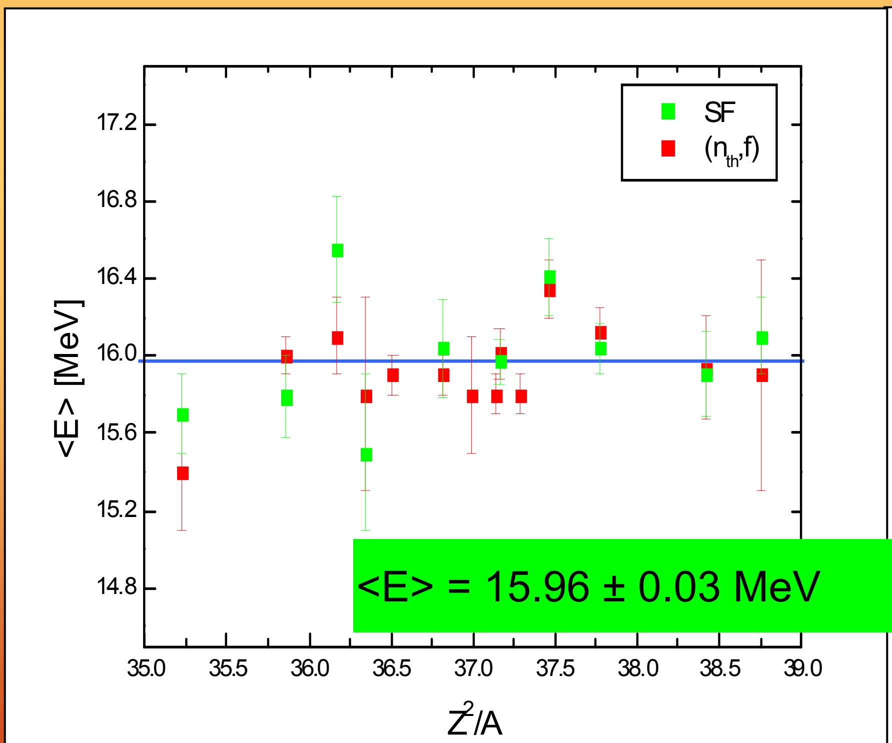
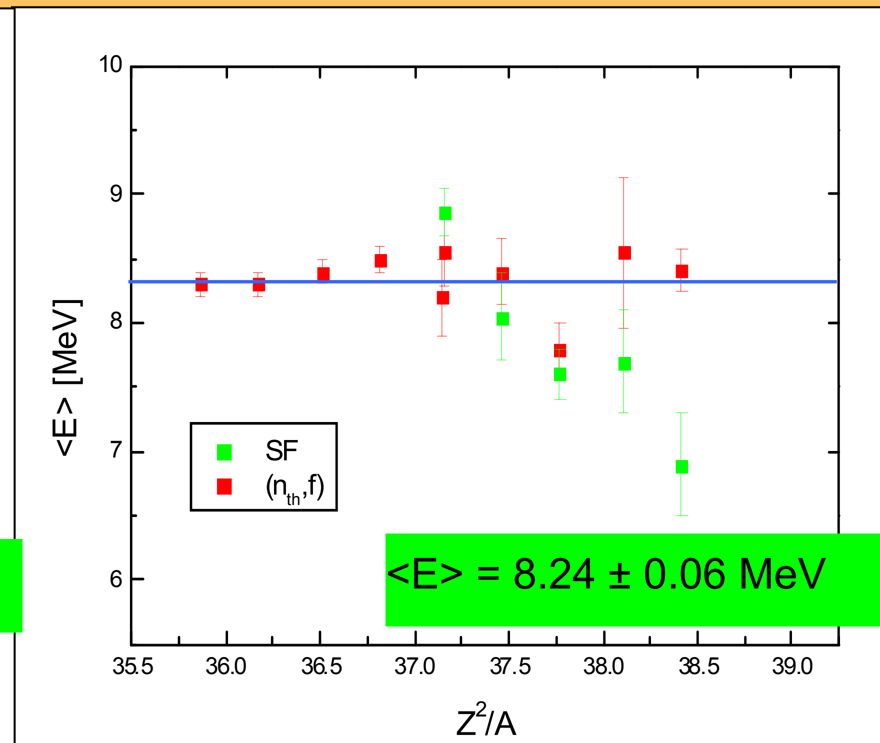
Average energy <E>

(as a function of the fissility parameter Z^2/A of the compound nucleus)

➡ remains practically constant for all fissioning systems

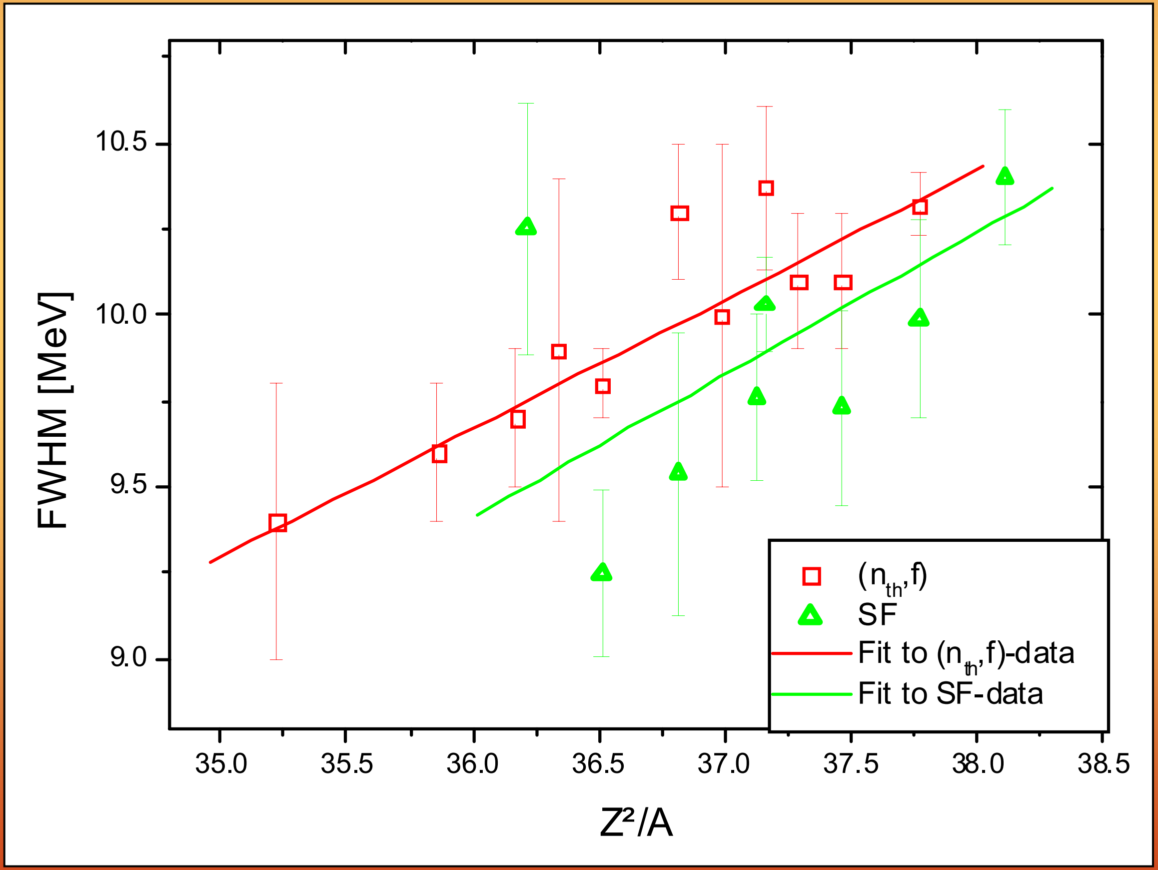




V | vwhp dwlf vwxg |

FWHM LRA

(as a function of the fissility parameter Z^2/A)



Increases with increasing Z^2/A

FWHM(SF) < FWHM(n_{th},f)

(excitation energy broadens)

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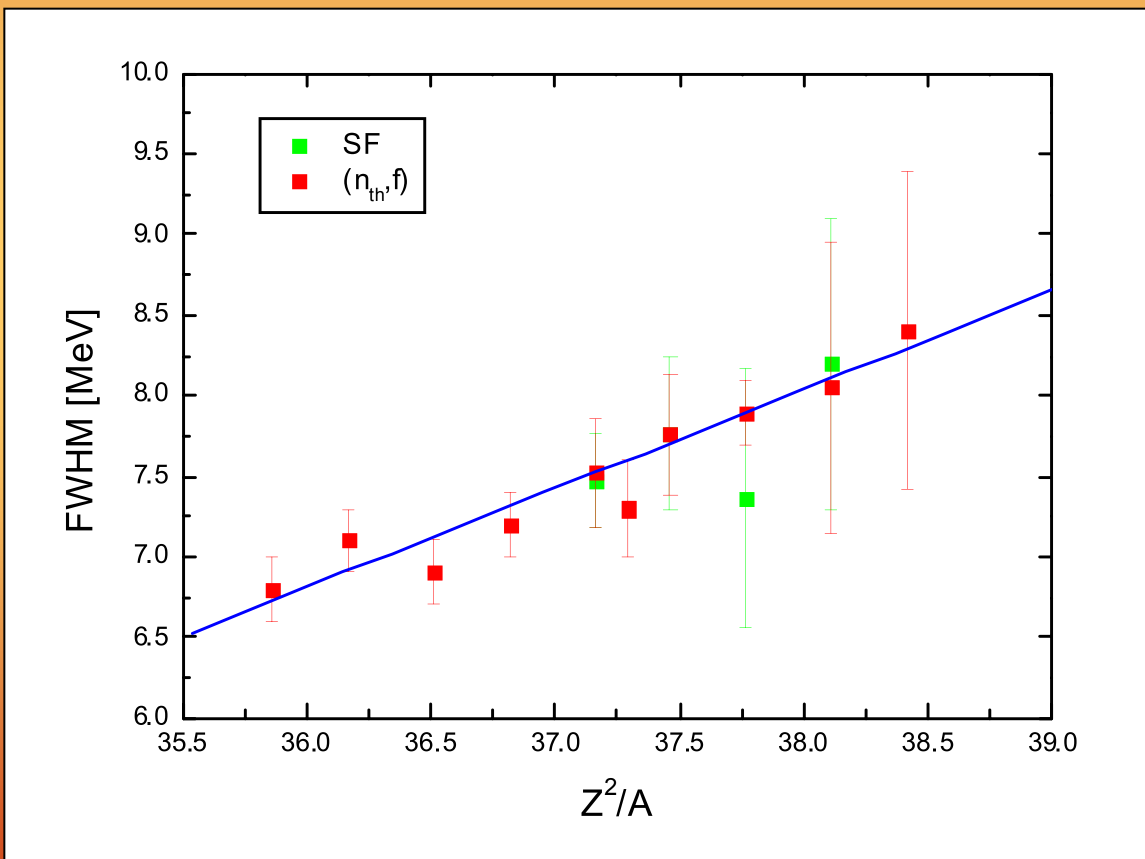
FWHM triton

(as a function of the fissility parameter Z^2/A)

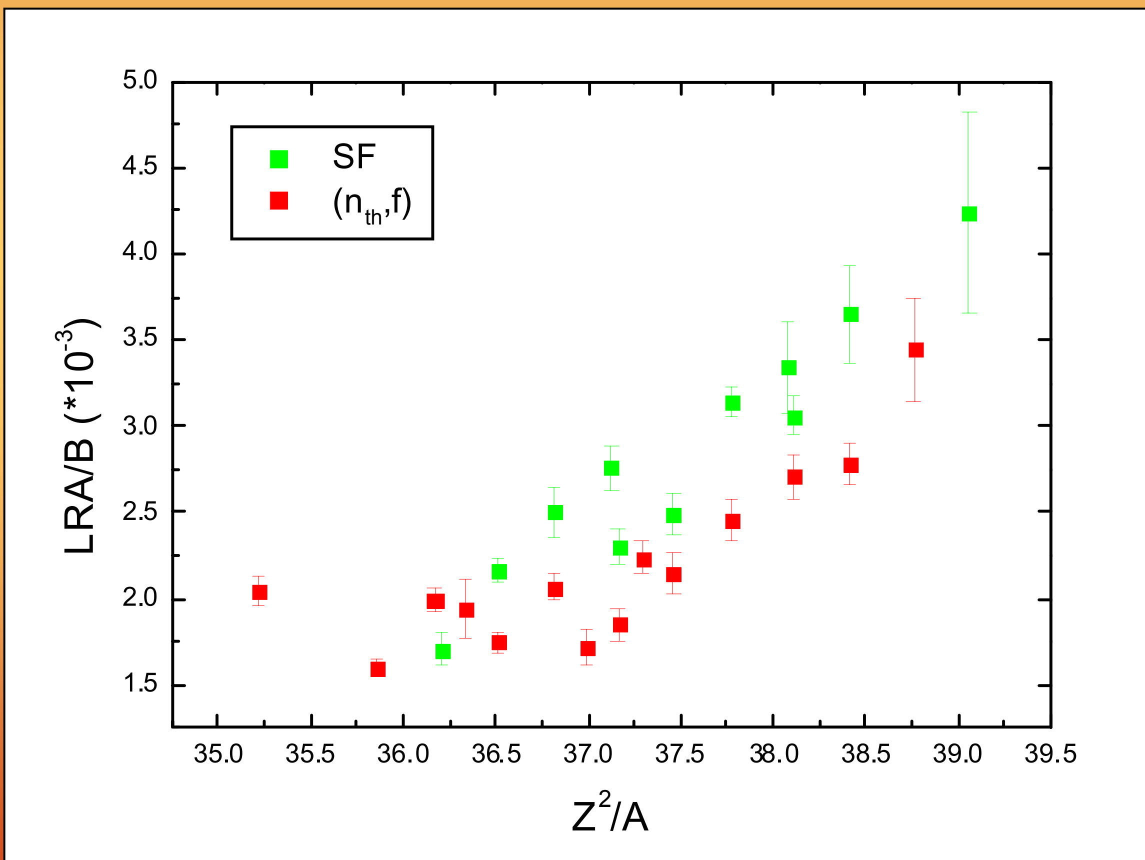


➡ Increases with increasing Z^2/A

V|vwhp dwlf vwxg|

Emission probability: LRA/B

(as a function of the fissility parameter Z^2/A)



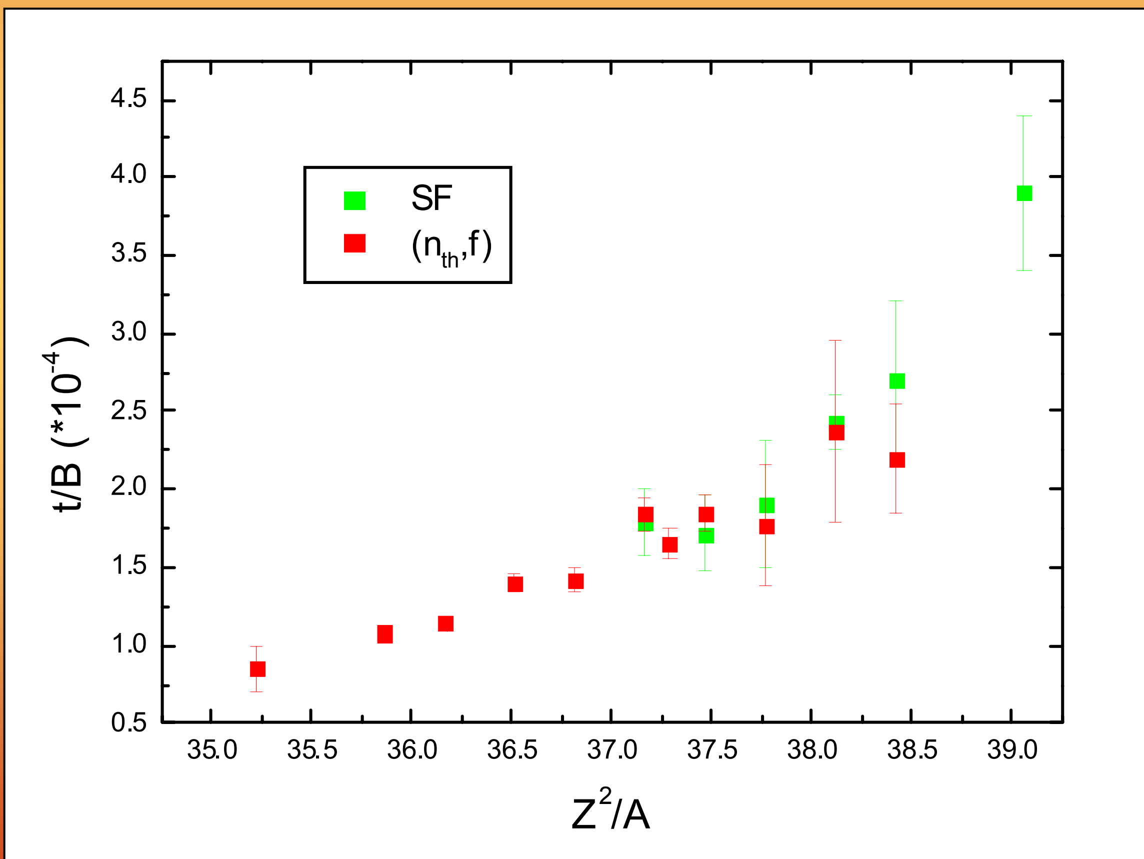
➡ Increases with increasing Z^2/A

(Z^2/A is a measure of the deformation)

V | vwhp dwlf vwxg |

Emission probability: t/B

(as a function of the fissility parameter Z^2/A)



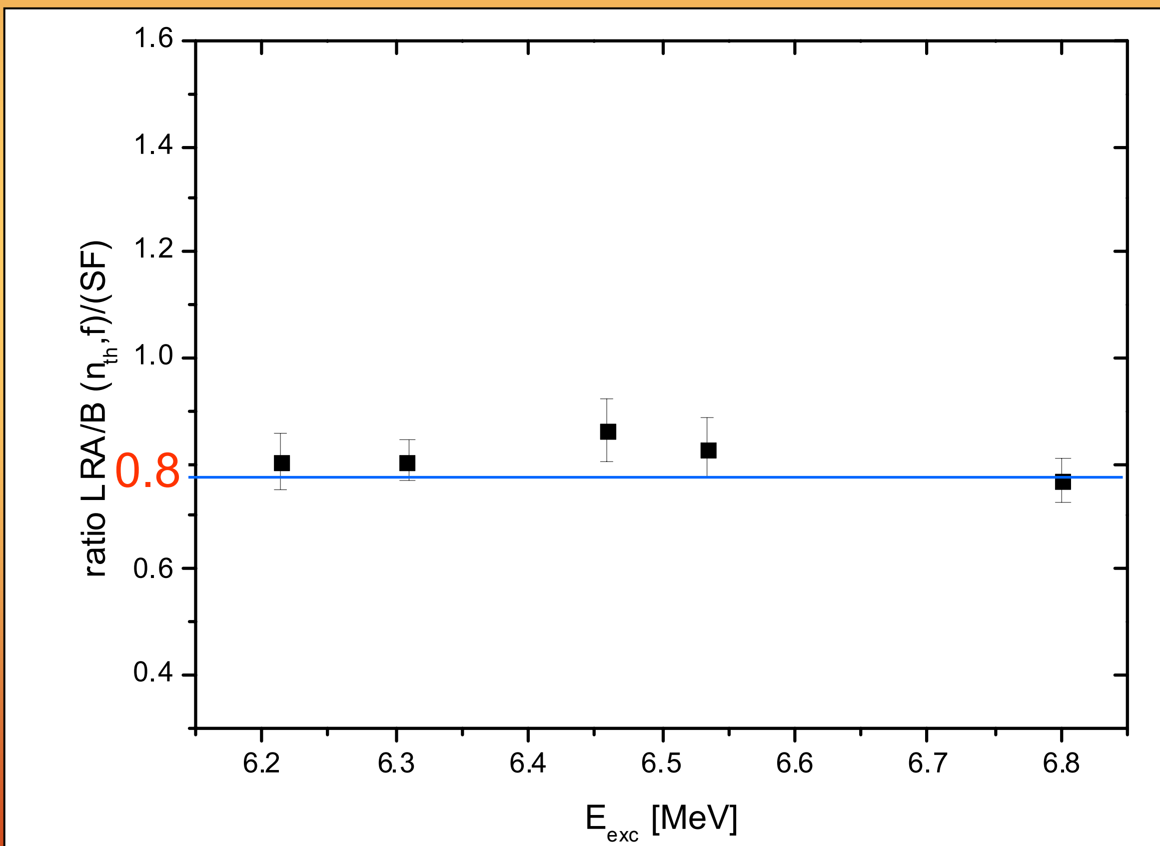
➡ Increases with increasing Z^2/A

(Z^2/A is a measure of the deformation)

V|vwhp dwlf vwxg|

Influence of the excitation energy E_{exc} on emission probabilities

➡ LRA: ^{240,242}Pu, ^{244,246,248}Cm

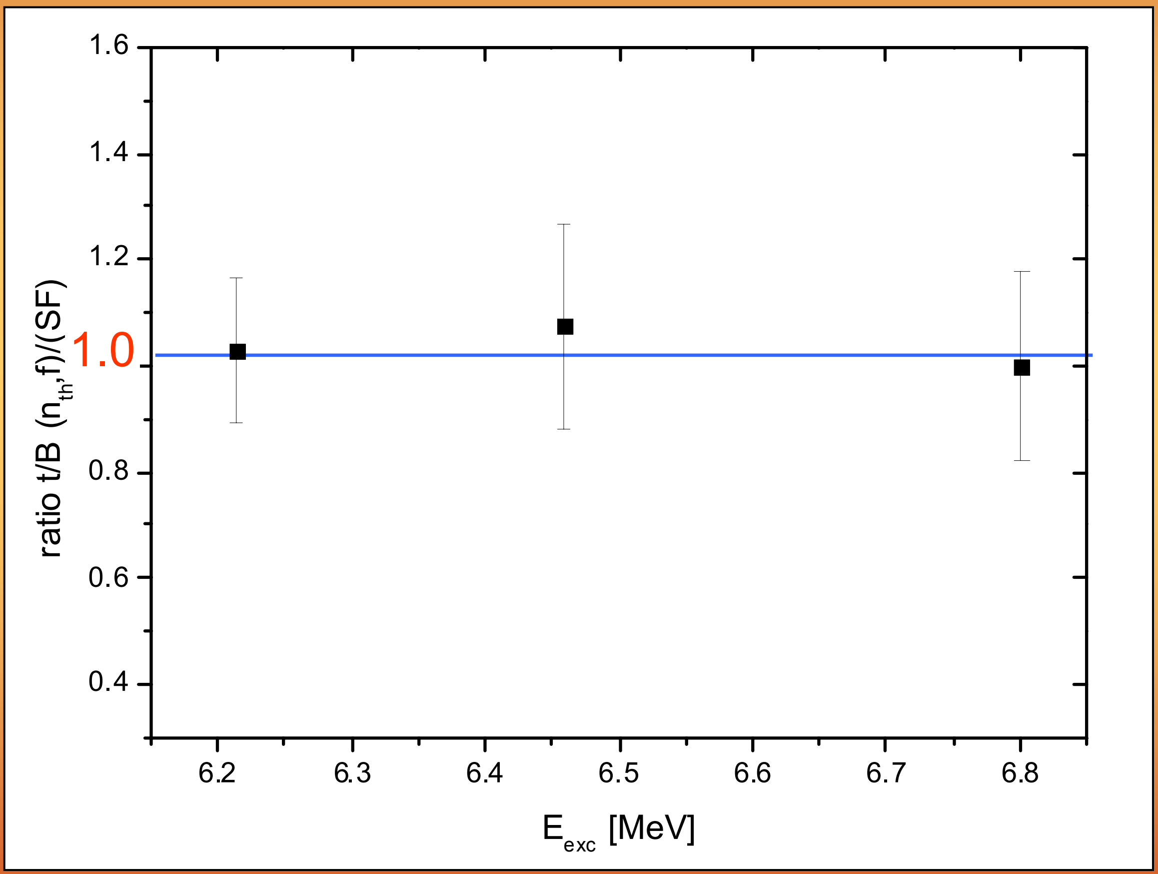


Ratio:

$$\frac{LRA/B(E_{exc})}{LRA/B(E_{exc}=0)}$$

V|vwhp dwlf vwxg|

➡ t: $^{244,246,248}Cm$



Difference between LRA and t

Influence of the factor S_α

(Alpha cluster preformation probability)

Decreasing value of S_α after capture of a neutron

This factor is not involved in the triton emission process

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Conclusions

- Experimental results on the ternary particle characteristics for ^{243,244}Cm and ^{249,251}Cf are presented
- The energy distribution characteristics (<E> & FWHM) can be well described in a systematic way
- Comparing the influence of the excitation energy on the emission probabilities for (SF) and (n_{th},f) a strong difference between t and LRA is observed

Outlook

- Further measurements on ^{6}He particles should be done in order to get an insight their behaviour
- Measurement on ^{250}Cf(SF) is just started, results will arrive